Electronic Structure Fermi Liquid Theory of High T_c Superconductors; Comparison of Predictions with Experiments

Jaejun Yu and A.J. Freeman
Department of Physics and Astronomy and Materials Research Center
Northwestern University, Evanston, IL 60208-3112

Abstract

Predictions of local density functional (LDF) calculations of the electronic structure and transport properties of high T_c superconductors are presented. As evidenced by the excellent agreement with both photoemission and positron annihilation experiments, a Fermi liquid nature of the 'normal' state of the high T_c superconductors become clear for the metallic phase of these oxides. In addition, LDF predictions on the normal state transport properties are qualitatively in agreement with experiments on single crystals. It is emphasized that the signs of the Hall coefficients for the high T_c superconductors are not consistent with the types of dopants (e.g., electron-doped or hole-doped) but are determined by the topology of the Fermi surfaces obtained from the LDF calculations.

INTRODUCTION

The most exciting issues in the microscopic theory of high T_c superconductivity are embodied in the questions: "What are the mechanisms of high T_c ?" and "What is the nature of the normal state of the Cu-oxide superconductors ?" A major issue is the understanding of the normal state of these systems, and, in particular, how well a Fermi liquid picture —in which for instance the quasiparticle energy band structure is approximated by the eigenvalues obtained within the local density approximation (LDA)— works in describing their normal state properties.

For years, there has been controversy and confusion among theorists as well as experimentalists on whether the 'normal' state of the Cu-oxide superconductors is a Fermi liquid or some other exotic ground state. Recently, however, some experimentalists (including Arko *et al.*[1]) are clarifying the nature of the normal state of the high T_c superconductors, surmounting the experimental difficulties in producing clean, well characterized surfaces so as to obtain meaningful high-resolution angle-resolved photoemission data. Their results agree with earlier positron-annihilation experiments[2]. Hence it has become apparent that, while the antiferromagnetic properties of the parent insulating compounds are not accounted for by present implementations of the local spin density approximation[3], the properties of these materials in the metallic region are well described by LDA band theory — as evidenced by the agreement between predictions of theory with both photoemission and positron annihilation experiments.

Recently, for $Nd_{2-x}Ce_xCuO_4$ systems, we obtained a positive Hall coefficient for the magnetic field oriented perpendicular to the Cu-O planes. This is to be compared with a negative experimental value found for $x < 0.18$ and recent experiments which show a change of sign of this Hall coefficient (from negative to positive with increasing x) for $x = 0.18$. Moreover, the measured Hall coefficient for the non-Cu based $Ba_{1-x}K_xBiO_3$ system was found to be negative, which agrees with our energy band results[4]. In addition, LDA predictions on the normal state transport properties for $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_7$ by Allen

et al.[5] These results on Hall coefficients indicate a trend (previously found for $La_{2-x}Sr_xCuO_4$) toward a regime where the conventional band theoretical description come into better agreement with experiment.

In this paper, we present the results of the local density functional calculations of the electronic structure and transport properties of high T_c superconductors and compare them with recent experiments. A Fermi liquid picture of the 'normal' metallic state of the high T_c superconductors is established through a detailed comparison of high-resolution angle-resolved photoemission spectra with our predictions of energy band dispersions and Fermi surfaces. In addition, we report a band theory based calculation of the Hall coefficients for cubic perovskite $Ba_{1-x}K_xBaO_3$ and for $Nd_{2-x}Ce_xCuO_4$ following those of Allen *et al.*[5] for the transport properties of $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_7$. The LDA predictions of the normal state transport properties are qualitatively in agreement with experiments on single crystals. Particularly it is emphasized that the signs of the Hall coefficients for the high T_c superconductors are not consistent with the types of dopants (e.g., electron-doped or hole-doped) but are determined by the topology of the Fermi surfaces.

LDA BAND STRUCTURE

For the electronic structure calculations, we used the highly precise full-potential linearized augmented plane wave (FLAPW) method [6] within the local density approximation (LDA) and the Hedin-Lundqvist form for the exchange-correlation potential. In the FLAPW approach no shape approximations are made to either the charge density or the potential. Results obtained on the high T_c Cu-oxides we studied — La_2CuO_4, $YBa_2Cu_3O_{7-\delta}$, $Bi_2Sr_2CaCu_2O_8$, $Tl_2Ba_2CaCu_2O_8$ and $Tl_2Ba_2Ca_2Cu_3O_{10}$ — indicate a number of common chemical and physical features, especially the role of intercalated layers such as the CuO chains, Bi_2O_2 and Tl_2O_2 rock-salt type layers. In the following, we provide a brief summary of the results on the electronic structure of $YBa_2Cu_3O_7$ as an illustrative example, compare them with experiments, and discuss its implication on the nature of the normal ground state of the high T_c Cu-oxides.

The calculated band structure[7, 8] of stoichiometric $YBa_2Cu_3O_7$ along high symmetry directions in the bottom ($k_z = 0$) plane of the orthorhombic Brillouin zone is shown in Fig. 1. As seen, a remarkably simple band structure near E_F emerges from this complex set of 36 Cu-O hybrid bands. Four bands — two each

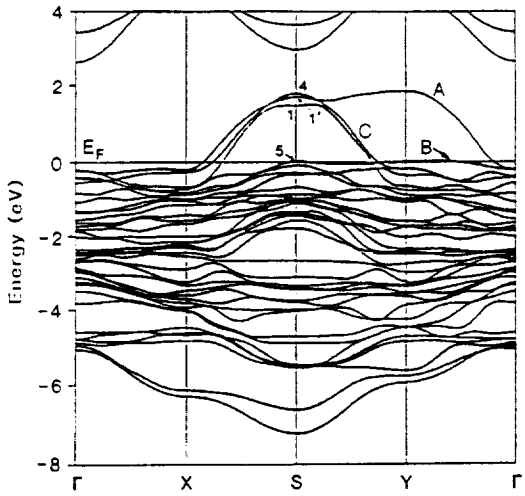

Figure 1: Band Structure of $YBa_2Cu_3O_7$ along symmetry directions in the $k_z = 0$ plane of the orthorhombic Brillouin zone

consisting of Cu(2) d - O(2) p - O(3) p orbitals and Cu(1) d - O(1) p - O(4) p orbitals — cross E_F. Two strongly dispersed bands labelled C ($S_{1'}$ and S_4 in Fig. 1; the labels are given by their character at S) consist of Cu(2) $d_{x^2-y^2}$ - O(2) p_x - O(3) p_y combinations and have the 2D character, which is also common in other high T_c Cu-oxide systems. Significantly, the Cu(1) $d_{x^2-y^2}$ - O(1) p_y - O(4) p_z anti-bonding band labelled A (S_1 in Fig. 1) shows the (large) 1D dispersion expected from the Cu(1)-O(1)-Cu(1) linear chains but is almost entirely unoccupied. This band is in sharp contrast to the π anti-bonding band labelled B (formed form the Cu(1) d_{xy} - O(1) p_z - O(4) p_y orbitals) which is almost entirely occupied in the stoichiometric ($\delta = 0$) compound.

In Fig. 2, our predicted Fermi surfaces (FS) of $YBa_2Cu_3O_7$ determined from our band structure are compared with experimental Fermi surfaces determined by high-resolution angle-resolved photoemission (ARPES) experiments by Campuzano *et al.*[9]. Filled circles indicate the points at which bands are found to cross the Fermi level. The open circles are the points in the Brillouin zone at which bands crossing E_F are not detected. (The size of the circles represents the experimental uncertainty in momentum.) The dashed lines are the predicted FS in Ref. [8]. Two 2D Cu-O $dp\sigma$ bands yield two rounded square FS's centered around S. It is remarkable that these 2D FS's have strong nesting features along the (100) and (010) directions, which are commonly found in other high T_c Cu-oxides except for the case of La-Sr-Cu-O. In addition, the 1D elec-

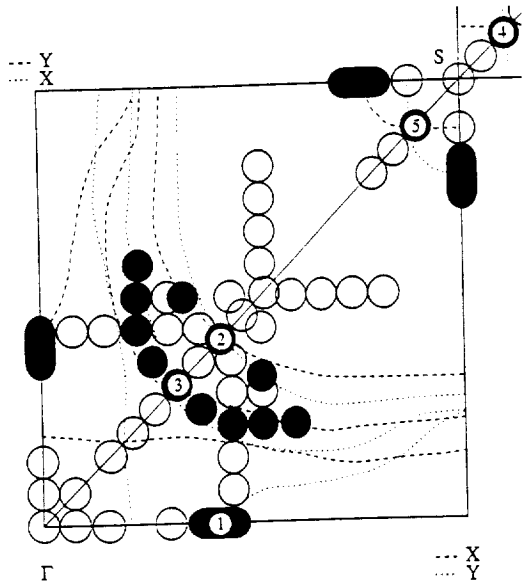

Figure 2: Comparison of the calculated Fermi surfaces with the experimentally determined Fermi surfaces of $YBa_2Cu_3O_7$. See the text for details. (This figure is taken from the paper by Campuzano *et al.*[9])

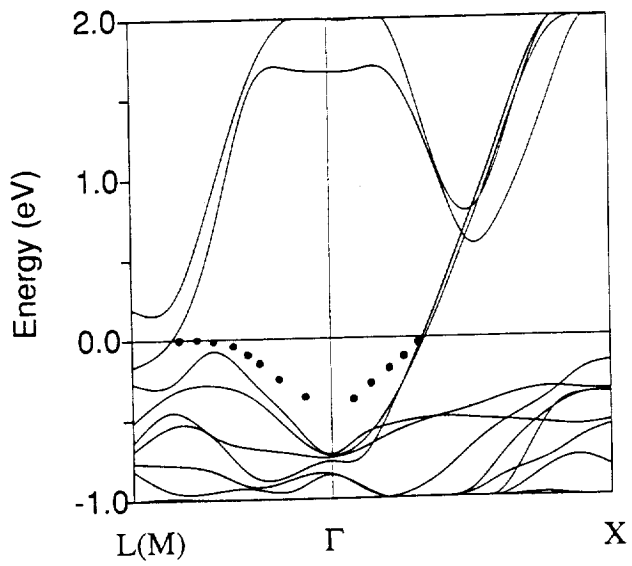

Figure 3: Comparison of the calculated energy bands with the measured bands in the ARPES experiments for $Bi_2Sr_2CaCu_2O_8$.

tronic structure also gives a 1D FS with again strong nesting features along the (010) direction. There are additional hole pockets around S(R) which come from the flat $dp\pi$ bands (band **B**) at E_F. In general, there is a remarkable agreement between the ARPES measurement of FS and our theoretical predictions on the FS of $YBa_2Cu_3O_7$. All of the predicted FS are observed in the experiment. Furthermore, there is also good agreement between the FS measured in the ARPES experiment and the FS obtained by positron annihilation experiment by Smedskjaer *et al.*[2]. These results demonstrate that the observed Fermi surfaces in $YBa_2Cu_3O_7$ are in agreement with LDA band calculations and consistent with the Luttinger theorem, where the large FS volume, which should be unaffected by the interaction, indicates the Fermi liquid nature of the normal ground state of these high T_c superconductors. It is important to note that the confirmation of the FS results has significant impact on several theories, which deny the Fermi liquid nature of the normal ground state in the Cu-oxide superconductors.

Another detailed ARPES study has been carried out on the $Bi_2Sr_2CaCu_2O_8$ system by Olson *et al.*[10] and leads to a good agreement with the LDA calculated band dispersions and Fermi surfaces. Fig. 3 shows a comparison of the calculated energy bands with the observed bands by Olson *et al.*[10]. (Filled circles in Fig 3 represent the observed band dispersions.) The agreement on the FS dimension between the experiment and the LDA band calculations is good, as also expected from the case of $YBa_2Cu_3O_7$. The observed band dispersions along major symmetry lines are large and indicate an enhanced effective mass, which may come from the renormalization effects due to electron-phonon interactions or Coulomb correlations.

The existence of Fermi surfaces and the large band dispersions observed in $YBa_2Cu_3O_7$ and $Bi_2Sr_2CaCu_2O_8$ together with such good agreement between LDA band theory and experiment strongly supports a Fermi liquid description of these high T_c superconductors.

NORMAL STATE TRANSPORT PROPERTIES

Normal state transport properties are of course an important test of the band theory of high T_c superconductors. The calculations based on the band theory have previously been performed for $La_{2-x}M_xCuO_4$ and $YBa_2Cu_3O_7$ by Allen *et al.*[5], and gave important results; these included a prediction of a change of sign in the current carriers at $x \approx 0.24$ for La-M-Cu-O, and the positive and negative signs for the Hall coefficients in $YBa_2Cu_3O_7$ when the magnetic field is aligned parallel and perpendicular to the c-axis, respectively. The

latter prediction has been confirmed by single crystal experiments[22], while the actual x value at which R_H changes sign is larger ($x \approx 0.3$) in experiments[11] on La-M-Cu-O.

An interesting test system for theory is the Ba-K-Bi-O system, for which it is a matter of discussion whether the conventional electron-phonon mechanism produces superconductivity, or if a different mechanism, possibly common to the cuprates, is necessary. In any event, if the K substitution into the insulating parent compound $BaBiO_3$ were regarded as hole-doping (i.e., if $Ba_{1-x}K_xBiO_3$ were a hole-doped superconductor), one would expect a positive Hall coefficient for the positive current carriers in $Ba_{1-x}K_xBaO_3$. Instead, the measured Hall coefficient (R_H) for the $Ba_{1-x}K_xBaO_3$ system was found to be negative[12, 13], which is in agreement with the calculated electron-like Fermi surface of this system[14] as shown in Fig. 4.

Another system obtained by electron doping of the parent compound, Nd_2CuO_4, is the recently discovered Nd-Ce-Cu-O superconductor[15]. In contrast to the case of $Ba_{1-x}K_xBiO_3$, if the Ce substitution into the insulating compound Nd_2CuO_4 were regarded as electron-doping, one would expect a negative Hall coefficient for the negative current carriers in $Nd_{2-x}Ce_xCuO_4$. There are, however, only few uncertain experiments available to date.

We present results in Tables 1 and 2 of a band theory based calculation of the Hall coefficients for cubic perovskite $Ba_{1-x}K_xBaO_3$ and for $Nd_{2-x}Ce_xCuO_4$ together with those of Allen *et al.*[5] for $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_7$. Our approach is based on the Bloch-Boltzmann theory, and rests on the Midgal approximation, where the sign of the Hall coefficients is mainly determined by the curvature of FS, i.e., the FS topology. Thus, one expects to have a positive R_H from the hole-like FS and a negative R_H from the electron-like FS, while the open FS can give rise to either positive or negative R_H depending on details of the FS geometry.

The Hall coefficient of the $Ba_{1-x}K_xBaO_3$ system in its cubic perovskite structure, as a function of K concentration, was calculated[4] within a rigid-band model (which was shown to be a good approximation for this system by a supercell calculation[16]). Our results are expected to reflect the experimental situation only in the region $x > 0.25$, since, as is well known, pure $BaBiO_3$ is an insulator and its structure has a monoclinic distortion. In the metallic region, the Hall coefficient is seen to be negative, and varies substantially with x. A negative sign of the Hall coefficient for $x > 0$ is, of course, expected from the electron-like Fermi surface of this system[14], as shown in Fig. 4.

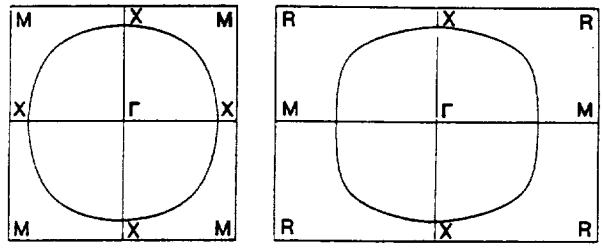

Figure 4: Fermi surface of $Ba_{1-x}K_xBiO_3$ ($x = 0.29$) within a rigid band model.

Experimentally, Sato *et al.*[12] measured a negative Hall coefficient in a single crystal, for $x \sim 0.39$ The measured R_H value ranges around $-4 \times 10^{-4}\text{cm}^3\text{C}^{-1}$ and with a weak temperature dependence. The sign of the measured R_H is in perfect agreement with our calculated electron-like FS, although the magnitude of the calculated R_H is larger than the measured R_H by a factor of ~ 2. Note, however, that this kind of disagreement in the magnitude of R_H between theory and experiment is not new. For instance, the calculations of Beaulac and Allen[17] showed that a factor of ~ 2 between theory and experiment exists for pure Pd if an isotropic scattering assumption is used, and that by contrast the inclusion of a moderate anisotropy gives good agreement with experiment.

The first thing to be pointed out for $Nd_{2-x}Ce_xCuO_4$ is the highly anisotropic nature of its transport properties. This is made apparent by comparison of the Fermi surface (FS) cuts on the basal and top planes of the Brillouin zone[18], plotted in Fig. 5 for x = 0.15. The mid-plane FS (not shown in Fig. 5 for clarity) lies in between those plotted. The striking closeness of these two curves illustrates the nearly vanishing dispersion along k_z of the Cu-O dps band near E_F. This feature is also common[19] to the $Tl_2Ba_2CaCu_2O_8$ system, for which a partial confirmation exists from preliminary positron annihilation measurements of the FS[20]. No such feature is found in $La_{2-x}Sr_xCuO_4$[21].

The calculated Hall coefficients for $Nd_{2-x}Ce_xCuO_4$ as a function of x (in a rigid band scheme) over a range including the superconducting region show, due to tetragonal symmetry, two independent components, $R_{xyz} = R_{yzx} \neq R_{xyz}$. R_{xyz} has a positive sign, as expected from the hole-like two-dimensional projection of the Fermi surface (cf. Fig. 5). On the other hand,

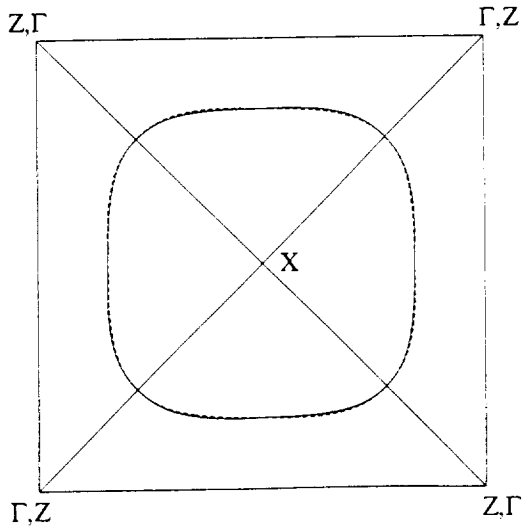

Figure 5: Fermi surface of $Nd_{2-x}Ce_xCuO_4$ ($x = 0.15$) plotted on the basal (solid line) and on the top (dashed line) planes of the Brillouin zone. A rigid band model is used.

this hole-like FS exists in $La_{2-x}M_xCuO_4$ only for a restricted range of $x < 0.24$[5, 21]. This difference relative to the "hole-analog" compound, La-M-Cu-O, is not surprising, since its Fermi surface[21] is quite different from that of $Nd_{2-x}Ce_xCuO_4$, and in particular shows a change of topology with compositions corresponding to the presence of a saddle point singularity in its band structure. The dependence of R_{xyz} on Ce concentration is rather weak.

The two other components ($R_{zxy} = R_{yzx}$) for $Nd_{2-x}Ce_xCuO_4$ are negative, and their magnitudes are comparable to that of R_{xyz}. A negative sign for these Hall coefficients, which correspond to the case of an in-plane magnetic field, is common to all the cuprate superconductors for which such calculations[5] have been performed, and was experimentally verified on single crystals of $YBa_2Cu_3O_7$ by Tozer *et al.*[22]. In fact, the magnetic field applied parallel to the Cu-O plane corresponds to the open topology of the zx-cross section of the Fermi surface.

Only a few experiments are available to date for the Nd-Ce-Cu-O system. In the semiconducting region ($x = 0.12$), single crystal measurements[23] showed a negative Hall coefficient, with a value consistent with the Ce concentration. Measurements on polycrystalline samples by Takagi *et al.*[24] showed that the charge carriers are electrons over a wide range of x values, but a departure from the $R_H \approx 1/n$ behavior was apparent. On the other hand, in the metallic (or superconducting) region ($x > 0.14$), recent measurements on single crystals showed interesting behavior of the Hall coefficients. The measurements by Takagi *et al.*[24] on polycrystalline samples and single crystals (with the magnetic field oriented parallel to the c-axis) showed a negative Hall coefficient but a change of sign (from negative to positive) for $x \approx 0.18$. Furthermore, the Hall coefficients measured by Ong *et al.*[25] are found to be positive for many superconducting samples of $Nd_{2-x}Ce_xCuO_4$ ($x \sim 0.15$).

Actually, this transition from the insulating ($x <\sim 0.14$) region, characterized by a negative Hall coefficient, to the metallic region, in which band structure effects should (according to our calculation) produce a positive R_{xyz} value, needs to be clarified from the theoretical point of view. The fact that experimentally a negative sign persists over the insulating and superconducting range might be due to an inhomogeneous Ce distribution over the sample, resulting in a clear departure of R_{xyz} from $1/n$ behaviour but still keeping a negative value. Further experimental work might clarify the situation.

A brief summary of both theoretical and experimental results on the Hall coefficients is given in Table 1 and 2. Note that the "doped-insulator" picture of high T_c superconductors is in disagreement with the experimental observations. On the other hand, the LDA band theory gives correct predictions on the signs of the Hall coefficients as determined by the FS topologies. For example, although the $Ba_{1-x}K_xBiO_3$ is regarded as "hole-doped", the Hall coefficient is not positive, but negative in agreement with the LDA calculations. Comparing the results of Hall coefficient measurements with the (p-type or n-type) "doped-insulator" picture, we are led to the conclusion that a separation of carriers into positive and negative in the spirit of the effective mass approximation is not possible for these copper oxides.

CONCLUSIONS

The energy band dispersions and Fermi surfaces observed in ARPES experiments are in good agreement with the predictions of LDA band theory. Further, the LDA predictions of the normal state transport properties are qualitatively in agreement with experiments on single crystals. In particular, it is emphasized that the signs of the Hall coefficients for the high T_c superconductors are not consistent with the types of dopants (e.g., electron-doped or hole-doped) but are determined by the local density functional calculations of the topol-

Table 1: Hall coefficients with **H** || **c**

	"doped" carrier	Fermi surface topology	band theory	experiment
$Ba_{1-x}K_xBiO_3$	hole	electron	− [a]	−
$Nd_{2-x}Ce_xCuO_4$	electron	hole	+ [a]	+(?)
$La_{2-x}Sr_xCuO_4$	hole	hole → electron	+ → − [b]	+ → −
$YBa_2Cu_3O_7$	hole	hole	+ [b]	+

[a] Ref. [4]
[b] Ref. [5]

Table 2: Hall coefficients with **H** ⊥ **c**

	"doped" carrier	Fermi surface topology	band theory	experiment
$Ba_{1-x}K_xBiO_3$	hole	electron	− [a]	−
$Nd_{2-x}Ce_xCuO_4$	electron	open	− [a]	(?)
$La_{2-x}Sr_xCuO_4$	hole	open	− [b]	−
$YBa_2Cu_3O_7$	hole	open	− [b]	−

[a] Ref. [4]
[b] Ref. [5]

ogy of the Fermi surface.

The existence of Fermi surfaces and large band dispersions observed in $YBa_2Cu_3O_7$ and $Bi_2Sr_2CaCu_2O_7$ together with such good agreement between LDA band theory and experiment strongly supports a Fermi liquid description of these high T_c superconductors. As a Fermi liquid (metallic) nature of the 'normal' state of the high T_c superconductors becomes clear, these experimental observations have served to confirm the predictions of our local density functional calculations and hence the energy band approach as a valid natural starting point for further studies of their superconductivity.

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